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Laser ablation of silicon dioxide on silicon using femtosecond near infrared laser pulses

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Abstract

Applying ultrashort laser pulses at wavelengths of 0.8 μm and 1.03 μm , the selective ablation of thin (~ 100 nm) SiO_2 layers from silicon wafers has been investigated. In particular, the effects of different pulse durations down to a minimum value of 50 fs, and single- as well as multi-pulse irradiation have been studied. Selective removal of the dielectric layer without any visible damage of the opened Si wafer was only possible with single pulse ablation. The threshold fluence for such complete ablation of the dielectric layer increases with increasing pulse duration. Irradiating two or more pulses on the same spot, significantly corrupted ablation craters are produced. The physical ablation mechanisms will be discussed with respect to the observed dependences on the laser pulse duration as well as on the number of laser pulses per spot.

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1. Introduction

The local removal of thin dielectric passivation and anti-reflection layers from silicon solar cells, e.g. for electrical contacts, by help of a flexible, mask-less laser ablation process has been the topic of several experimental studies in the last few years. It could be shown that intense laser pulses using photon energies above the energy gap of silicon can provide the desired ablation, apparently without considerable energy input in the transparent layer [1-3]. Also, it became clear rather quickly that nanosecond pulses cause much more thermally induced collateral damage in the silicon than pulses with durations of typically 10 picoseconds [4]. Very recently, we were able to show that the threshold fluence for such selective ablation drops further when the pulse durations are further decreased into the femtosecond range [5], accompanied by a wider fluence range allowing really damage-free ablation. However, this study was done using a Ti:sapphire laser system at 800 nm, while Yb-based ultrashort lasers currently appear as

more promising with respect to industry-relevant parameters like average power, repetition rate or stability. Therefore we report here novel selective ablation experiments obtained by help of an Yb:KGW laser operating at 1.03 μm wavelength with typical pulse duration of 280 fs, and compare them with the results obtained using femtosecond pulses at 800 nm. In addition, the effects of irradiating more than one pulse on the same spot are demonstrated and used to discuss the physical ablation mechanism.

2. Experimental

We have examined the local removal of approximately 100 nm thick thermally grown SiO_2 layers from planar silicon substrates. The ablation experiments have been conducted using different laser systems: (1) The Ti:sapphire laser systems Spitfire (Spectra Physics) with pulse duration of 50 fs and Legend (Coherent) with pulse duration between 0.7 ps and 2 ps, both at a wavelength of 0.8 μm . (2) The Yb:KGW laser system Pharos (Light Conversion) with a pulse duration of 280 fs and a wavelength of 1.03 μm . All laser beams had been focused on the sample surface by help of plano-convex lenses; the characterization of beam size and shape in the focal range was done by a uEye camera having a pixel size of 6 μm . The pulse energy was measured with a pyroelectric sensor. Fine tuning of the pulse energy was done in two ways: (1) with a computer controlled half-wave plate and a thin-film polarizer using the Ti:sapphire laser systems and (2) by a software controlled change of the regenerative amplifier current combined with a photodiode monitored readjustment (Pharos).

The substrates were fixed with a vacuum-chuck on top of a motorized x-y-table; the precise movement of this positioning stage as well as the laser pulse sequence was computer controlled. To allow a reliable statistical evaluation, lines of 100 areas with a spot-to-spot distance of 100 μm and constant pulse energy have been prepared using this setup. For single-pulse irradiation at $\lambda = 0.8 \mu\text{m}$, different pulse energies were examined in different lines (also at a distance of 100 μm). The ablation experiments at $\lambda = 1.03 \mu\text{m}$ have been grouped in a different way to assure identical conditions for irradiating different numbers of pulses on the same area: groups of four lines were produced in the above described way using identical pulse energy, but an increasing (from line to line) number of 1, 2, 5 or 10 pulses per spot. Here, different pulse energies were examined in different blocks, which had a distance of 200 μm . After irradiating the samples, the machined areas were characterized by light microscopy (using a Zeiss Axioplan 2 imaging microscope) as well as atomic force microscopy (Witec alpha 300).

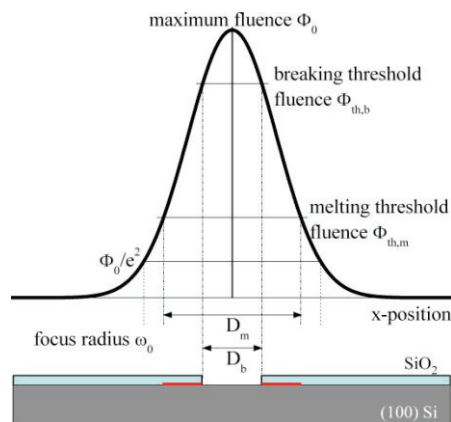


Fig. 1. Fluence profile of a Gaussian beam with beam radius ω_0 at Φ_0/ϵ^2 . Above the melting threshold fluence a change of colour (reflectivity) can be seen. Fluences higher than the breaking threshold fluence are necessary to ablate the SiO_2 layer on silicon substrate.

In accordance with previous work [5] we define two different threshold fluences: (i) The breaking threshold fluence $\Phi_{th,b}$ is the minimum local fluence needed for an ablation of the SiO_2 layer on the silicon substrate. (ii) Already at lower fluences visible colour (reflectivity) changes can be seen; we define the minimum fluence where such an effect is observed as the melting threshold fluence $\Phi_{th,m}$. Fig. 1 shows schematically where the two thresholds can typically be found within a Gaussian beam, and to which diameters D_m and D_b they are related.

3. Results

We have performed several series of ablation experiments on SiO_2 covered Si wafers, varying the irradiation parameters: pulse duration, number of pulses, and total energy density applied to one spot of the sample. In the following we will concentrate on the results obtained by irradiation at $1.03 \mu\text{m}$, which have all been done at the same pulse duration of ~ 280 fs.

All areas irradiated with a single pulse have shown the same qualitative behavior: Ordered from lower to higher laser fluence, one observes in the irradiated spots (a) irreversible modifications without opening of the SiO_2 surface (Fig. 2a), (b) SiO_2 ablation without visible damage of the Si wafer (Fig. 2b), and finally (c) SiO_2 ablation with modification of opened Si surface (not shown in the Figure). All topological modifications are surrounded by a so-called corona, i.e. a region in form of an annulus which is clearly visible in the optical microscope, but without measurable surface modification. These phenomena are more or less identical to those observed upon irradiation of single femtosecond pulses at $0.8 \mu\text{m}$ wavelength, which have been described in detail in our recent publication [5].

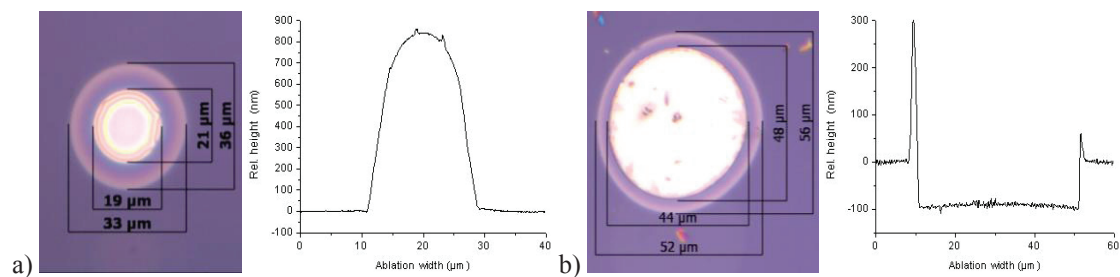


Fig. 2. Spots irradiated with single pulses (wavelength $1.03 \mu\text{m}$) of different fluences, measured with light microscopy (left) and AFM (right): (a) 100 mJ/cm^2 (b) 200 mJ/cm^2 .

If, however, two or more pulses were irradiated on the same spot, a quite different behavior emerged. Let us first discuss the case of five or more successive pulses on one area. Under these conditions, a selective, damage-free ablation recognizable by a flat crater bottom is apparently not possible at all: either, at lower laser fluences, there is no modification at all, or whenever one observes ablation at higher laser fluences, the opened silicon surface is completely ragged, obviously due to molten and re-solidified silicon. The case of two pulses per spot turns out to be much more interesting: as one could expect, ablation is being observed already at a roughly two times lower energy density per pulse, i.e. at the same total energy density like in the single pulse case. However, the ablation crater does not have the same principal appearance of flat opened Si surface surrounded by a corona, like in the single pulse case, but instead the ablated area is divided into two parts by a ring-like structure. These phenomena can nicely be seen in the microscope images shown in Fig. 3a and 3b, where the spots on the left-hand side of each image have been produced by applying only one pulse, those on the right-hand side by two pulses of the same energy (as specified in the figure caption). The comparison of singly and doubly irradiated areas at

the two different pulse energies gives a clear idea about the origin of this structure, since the ring in the two-pulse case appears to be located more or less exactly at the position where the border between bulge and corona was after one pulse.

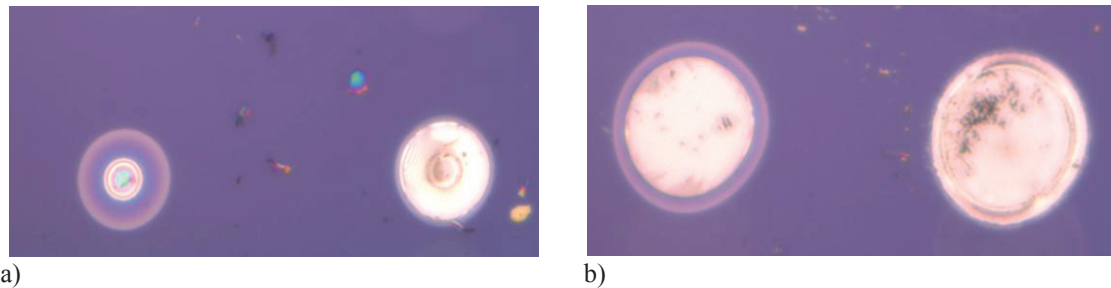


Fig. 3. Microscope images of spots irradiated with one (left-hand side) or two (right-hand side) pulses (wavelength 1.03 μm) of identical fluence per pulse: (a) 90 mJ/cm^2 (b) 180 mJ/cm^2 .

More details about the background of the visible ring after ablation with two successive pulses can be recognized in the AFM scan presented in Fig. 4b. Clearly, the ring is due to a wall with a height in the order of the thickness of the ablated SiO_2 layer (100 nm), and the opened silicon surface inside the ring is looking quite rough in contrast to the at least optically smooth outer annulus of the ablated region.

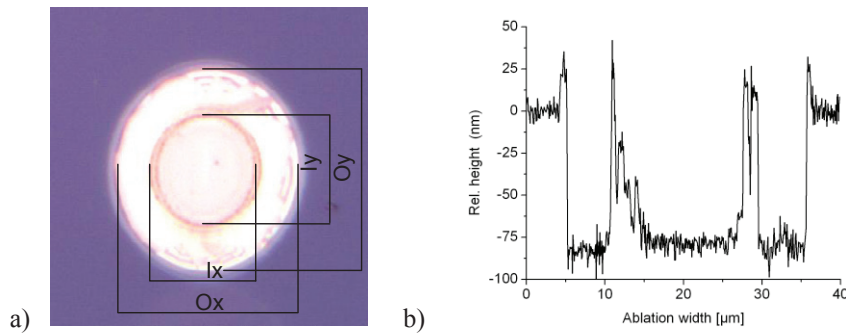


Fig. 4. Ablation area after two pulses measured by light microscopy (a) and AFM (b); inner ablation area $I(x,y)$ and outer ablation area $O(x,y)$ separated by an inner circle (wall)

We have measured as a function of pulse energy the diameters of ablation area and corona after one pulse ablation (squares and circles, respectively, in Fig. 5a), as well as the diameters of inner ring and total ablation area of the spots irradiated with two pulses (Fig. 5b). As can be clearly seen comparing the one- and two-pulse results in Fig. 5a and b, the diameters of ablated area and corona in the first case are in very good accordance with the diameters of inner ring and total ablated area in the case of two pulses. The measured dependences of the different diameters on the laser pulse energy can be used to determine the fluence thresholds for melting of silicon at the interface and ablation (breaking) of the dielectric layer both for one-pulse and two-pulse ablation. Using the description of Liu [6], the diameter of an irreversibly changed round area can be related to focus radius ω_0 , threshold fluence Φ_{th} and maximum fluence Φ_0 of the applied pulses by:

$$D^2_{(m,b)} = 2 \cdot \omega_0^2 \cdot \ln \left(\frac{2 \cdot E_{pulse}}{\pi \cdot \omega_0^2 \cdot \Phi_{th,(m,b)}} \right) \quad (1)$$

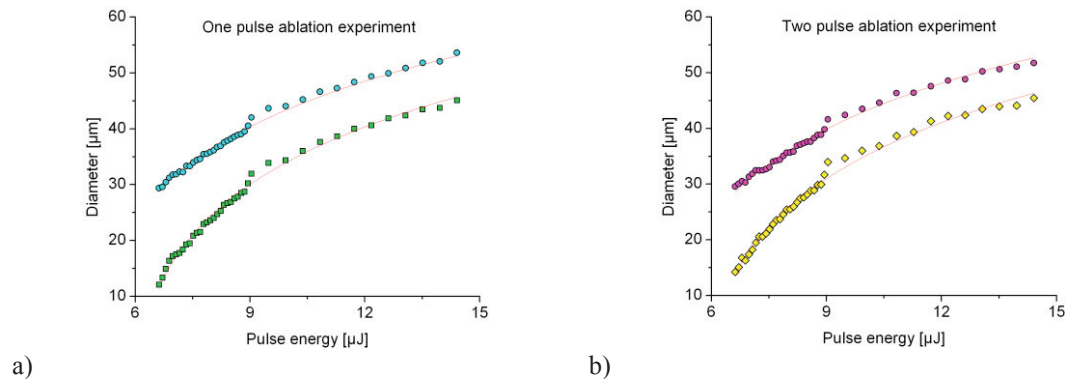


Fig. 5. Diameter of (inner) ablation (square) and corona / outer ablation (circle) after irradiation with (a) one pulse and (b) two pulses; solid curve refer to fits obtained using (1)

Least squares fits of our data based on Eq.(1) are presented in Fig. 5 as solid lines, showing that the description fits well (the deviation around a pulse energy of 9 μJ are apparently due to unnoticed laser energy fluctuations which occurred during the experiments). The threshold fluence obtained by this procedure for ablation after one pulse is 318 mJ/cm² and in a good agreement with the value of 311 mJ/cm² obtained for the inner ring observed after two pulses.

The same accordance was found for the diameter of the corona (238 mJ/cm²) and the diameter of the total ablated area after irradiation of two successive pulses (242 mJ/cm²). The focus radius was obtained consistently for all cases to be $\omega_0 = 35.5 \pm 0.5 \mu\text{m}$.

4. Interpretation and discussion

For the interpretation of the single-pulse ablation results with femtosecond pulses at $\lambda = 1.03 \mu\text{m}$ presented above it is instructive to compare them to the previously published results obtained at $\lambda = 800 \text{ nm}$ [5]. There, qualitatively identical phenomena were monitored, and a significant dependence of melting and breaking threshold fluences on the laser pulse duration was observed. An interpolation of the pulse duration experiment done with $\lambda = 800 \text{ nm}$ yields values of 205 mJ/cm² and 280 mJ/cm² respectively for a pulse duration of ~280 fs. This fluences are typically 15% lower than the values obtained here with the Yb:KGW laser.

This is well compatible with the idea that the ablation process in case of ultrashort laser pulses is by no means governed by the energy input due to linear or two-photon absorption based in equilibrium parameters, but is a highly nonlinear, dynamical process where the laser pulse itself generates an electron-hole plasma which is then responsible for strongly increased, quasi metallic absorption of most of the pulse energy. In this picture the linear absorption of Si, being smaller by more than an order of magnitude at $\lambda = 1.03 \mu\text{m}$ compared to 0.8 μm, only provides a different starting condition to reach a sufficiently high carrier density close to the Si/SiO₂ interface. This explains nicely why the thresholds are very similar in spite of the vastly different equilibrium extinction at the two different wavelengths, and also why the qualitative appearance of the ablated regions is basically identical using the two different laser wavelengths.

The results obtained with more than one ultrashort laser pulse on the same sample spot confirm that the selective, damage-free ablation of the dielectric layer is a very special situation which requires ultrafast input of the needed energy as well as a smooth, undisturbed interface from Si to dielectric layer.

It is true that nearly the same ablation threshold fluence is found when the total energy is irradiated in form of one or in form of two successive femtosecond pulses. However, while in the first case a smooth and clean opened Si surface surrounded by a corona is produced, the second case shows a ring wall within the ablation area, which has obviously been set up already by the interaction of the first pulse. More generally spoken, as soon as there is any irreversible change at this interface due to a first pulse (which itself is not strong enough to cause ablation), the interaction of the second pulse leading to ablation is considerably changed. In particular, even where the first pulse has produced ‘only’ a corona, the second one can ablate the material at lower energy density compared to the undisturbed interface. Turning the argument around, this is a clear confirmation of the idea that the corona is due to molten and re-solidified material, providing in some sense a predetermined breaking point facilitating ablation for a second pulse.

On the other hand, this insight defines an important constraint for technological application of this technique: if really damage-free opening of larger coherent areas on a Si wafer covered by a dielectric layer is intended, the overlap regions of successive pulses have to be constrained to the absolute minimum.

Summary

We have studied the selective ablation of thin (~ 100 nm) SiO_2 layers from silicon wafers by using ultra-short laser pulses at wavelengths of $0.8\ \mu\text{m}$ and $1.03\ \mu\text{m}$. Using different pulse durations between 50 fs and 2000 fs, we were able to show that the energy input is a highly dynamical process governed by carriers injected by the fs pulse itself. Therefore the threshold for the selective ablation process shows almost no dependence on the linear absorption coefficient of Si, but drops considerably towards shorter pulses duration in the femtosecond range. The other important finding of this work is that a flat selective ablation irradiating two or more pulses to the same area is not possible. Using five or more pulses there is complete melting and destruction of the opened silicon surface whenever ablation is observed at all. Even after irradiating only two successive pulses on one spot, the ablated area is disturbed by a ring-like wall structure; the diameter of this inner ring corresponds to the diameter of the ablated region after one pulse ablation, while the outer diameter of two-pulse ablation corresponds to the width of the corona observed after one pulse.

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